

Emergence of linear elasticity from the atomistic description of matter

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We investigate the emergence of the continuum elastic limit from the atomistic description of matter at zero temperature considering how locally defined elastic quantities depend on the coarse graining length scale. Results obtained numerically investigating different model systems are rationalized in a unifying picture according to which the continuum elastic limit emerges through a process determined by two system properties, the degree of disorder, and a length scale associated to the transverse low-frequency vibrational modes. The degree of disorder controls the emergence of long-range local shear stress and shear strain correlations, while the transverse length scale influences the amplitude of the fluctuations of the local elastic constants.

I. INTRODUCTION

The mechanical properties of a solid can be described by taking into account the discrete nature of matter, via the introduction of an equivalent network of masses and springs. If the size of the solid is much larger than the typical interatomic spacing, matter can be treated as a continuum and the mechanical properties are conveniently described through elasticity theory [1]. Here we consider how the continuum description sets it as the elastic properties of a material are investigated on increasingly larger spatial scales. This is a question easily addressed in ideal crystals, where the two descriptions can be related exploiting the periodicity of the lattice structure. [1, 2] In real crystals, that are made of many randomly oriented ideal micro crystals, periodicity is lost and the connection between the two descriptions is not obvious; in this case, one tries to recover the continuum description considering how the micro crystals rotate in response to external deformations [3, 4]. In amorphous materials the problem is more challenging, as disorder induces non-affine particle displacements [5] in response to macroscopic deformations [5]. It is known that this non-affine displacement lowers the elastic constants by influencing the fluctuation term of the stress tensor [6, 7]. However, the role of disorder on the emergence of the continuum limit is unclear. To investigate how disorder influences the emergence of linear elasticity, one needs to consider the dependence of locally defined elastic properties on the coarse-graining length scale, in systems with different degree of disorder. Numerically, the measure of local elastic properties can be carried out exploiting the derivation of exact coarse graining expression for local strain and stress fields [8–10], or via fluctuation formulas [11, 12]. In this research direction, previous results on a two-dimensional weakly polydisperse Lennard-Jones (LJ) [13] system, as well as on systems of

spheres interacting via finite range purely repulsive potentials [14], have found the standard deviation (s.d.) of elastic constants defined on a coarse graining length scale w to scale as $w^{-\alpha}$, with $\alpha = d/2$ in both [13] $d = 2$ and $d = 3$ [15] spatial dimensions. The value $\alpha = d/2$ has been rationalized through the central limit theorem, assuming local elastic constants to be the average of $\propto w^d$ random variables associated to the coarse graining region. However, a local modulus is not the sum of random variables, but rather the ratio of two coarse-grained quantities, a local stress change and a local strain change, so that the applicability of the central limit theorem is questionable. In addition, the central limit theorem holds in the presence of short range correlations, while the local shear strain could be long-range correlated, as observed in colloidal [16] and metallic [17] glasses, and theoretically predicted in granular systems [18]. These considerations suggest that the exponent α , and therefore the process by which continuum elasticity sets in, could not be universal.

Here we show, through the numerical investigation of different model systems with different degree of disorder, that the process by which continuum elasticity sets in is not universal. Rather, it depends on the local order and on a length scale associated to the transverse acoustic modes. The degree of local order fixes the value of the exponent α_G governing the asymptotic scaling of the s.d. of the local shear modulus G with the coarse-graining length scale, $\Delta G \propto w^{-\alpha_G}$. We find $\alpha_G = d/2$ only in the presence of a high degree of local order, as in previously investigated weakly polydisperse [13] or monodisperse systems [14]. In general, α_G depends on the degree of local order, and its value is understood from the coarse-graining length dependence of the s.d. of the local shear stress and of the local shear strain. The transverse vibrational modes fix the length scale ξ_G above which ΔG scale as a power law with w . In finite range purely repulsive particle systems this length scale diverges in the zero pressure limit, as the jamming transition is approached.

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II. LOCAL ELASTIC CONSTANTS

A. Definitions of Local Quantities

We use the approach developed by Goldhirsch [19] to define the local stress and strain based on coarse grained displacement fields. Local stress [8] and displacement fields [10] are defined as

$$\sigma_{\alpha\beta}(\mathbf{r}) = \frac{1}{2} \sum_{i \neq j} F_{ij\alpha} r_{ij\beta} \int_0^1 ds \phi_W(\mathbf{r} - \mathbf{r}_i + s\mathbf{r}_{ij}) \quad (1)$$

and as

$$\mathbf{u}^\ell(\mathbf{r}) = \frac{\sum_i m_i \mathbf{u}_i \phi_W(\mathbf{r} - \mathbf{r}_i)}{\sum_j m_j \phi_W(\mathbf{r} - \mathbf{r}_j)}. \quad (2)$$

Here \mathbf{u}_i is the displacement of particle i induced by an external deformation, and $\phi_W(\mathbf{r}) = \frac{1}{\pi W^2} \exp[-\mathbf{r}^2/W^2]$ is a 2d Gaussian coarse graining function [19]. $F_{ij\alpha}$ and $r_{ij\alpha}$ are the α components of contact forces between interacting particles i and j , $r_{ij} = |\mathbf{r}_{ij}| = |\mathbf{r}_i - \mathbf{r}_j|$ is the interparticle separation, m_i is the particle mass. Given the displacement field, the strain $\epsilon_{\alpha\beta}(\mathbf{r})$ is derived as

$$\epsilon_{\alpha\beta}(\mathbf{r}) = \frac{1}{2} \left(\frac{\partial u_\alpha^\ell(\mathbf{r})}{\partial r_\beta} + \frac{\partial u_\beta^\ell(\mathbf{r})}{\partial r_\alpha} \right). \quad (3)$$

We have used the above expressions to determine the local elastic properties on the points of a square grid of lattice spacing w , fixing the standard deviation of the coarse graining function to $W = w/5$, as in previous studies [20]. In the following, we consider w to be our coarse-graining length scale.

B. Standard Deviations

It is useful to recapitulate how the s.d. of different locally defined elastic quantities are expected to scale with the coarse graining length scale. Eqs. 1 and 3 clarify that the local strain and the local stress are averages of properties of contacts and of particles found within the coarse graining region, whose number scales as w^d . According to the central limit theorem their s.d should scale as $w^{-d/2}$ for w larger than their characteristic correlation length. Conversely, a local modulus is the ratio of a stress X and of a strain Y , $M = X/Y$. In general, there is not a closed form for the s.d. of the ratio of two random variables. However, if the probability distributions of Y is such that $P(Y = 0) \simeq 0$, then

$$\Delta_M^2 = \frac{\langle X \rangle^2}{\langle Y \rangle^2} \left[\frac{\Delta_X^2}{\langle X \rangle^2} + \frac{\Delta_Y^2}{\langle Y \rangle^2} - 2 \frac{\Delta_{XY}}{\langle X \rangle \langle Y \rangle} \right] \quad (4)$$

where $\langle X \rangle$ and $\langle Y \rangle$ are the average values of X and Y , and Δ_{XY} their covariance.

III. NUMERICAL MODELS

We investigate the emergence of linear elasticity in 2d LJ systems, that model atomic systems, and in 2d Harmonic particles, that model colloids, foams and granular matter, via zero temperature molecular dynamics simulations [21]. We consider two LJ systems. The first one is a mixture of N_L large and $N - N_L$ small particles, with different fraction $f_L = N_L/N$ of large particles in the range 0–1. We vary f_L to control the degree of disorder, as the size ratio of the two components is such that when $f_L \simeq 0$ ($\simeq 1$), our system is polycrystalline, while for $f_L \simeq 0.5$ it is amorphous. As second LJ system we consider the weakly polydisperse system studied in Ref 13, to rationalize previous results within our approach. We explore the role of the length scale associated to transverse vibrational modes considering Harmonic particles at different values of the pressure, as this length scale is known to diverge in the zero pressure limit [22]. We notice that previous studies have considered the dependence of the macroscopic elastic constants on disorder by tuning the relative fraction of different species [23]. However, the role of disorder on the emergence of the continuum limit has not been previously addressed.

A. Lennard Jones systems

In LJ systems particles interact with the potential

$$V(r_{ij}) = 4\epsilon \left[\left(\frac{d_{ij}}{r_{ij}} \right)^{12} - \left(\frac{d_{ij}}{r_{ij}} \right)^6 + V_{\text{offset}} \right] \quad \text{for } r_{ij} < r_c$$

$$V(r_{ij}) = 0 \quad \text{for } r_{ij} \geq r_c,$$

where $\epsilon = 1$ is the energy parameter, r_c is a cutoff, V_{offset} is such that $V(r_c) = 0$, $d_{ij} = d_i + d_j$, where d_i is the ‘diameter’ of particle i . We consider a bidisperse and a weakly polydisperse system. In bidisperse systems, large and small particles have a diameter ratio 1.4, and $r_c = 2.5d_{ij}$. In polydisperse systems, particle diameters are drawn from a uniform distribution with average value $\langle d \rangle$ and small s.d. $\sim 0.1\langle d \rangle$, so that the system is weakly disperse [13], and the cutoff $r_c = 2^{1/6}d_{ij}$ so that the interaction is purely repulsive. The systems are prepared at zero temperature minimizing the energy of interaction through the conjugate-gradient algorithm, starting from infinite temperature states, and have a pressure of the order of ten. For each set of parameters, our data are obtained averaging over 100 independent configurations with $N = 10^4$ particles.

B. Harmonic particle systems

In Harmonic systems, particles interact via the potential

$$V(r_{ij}) = \frac{1}{2} k (d_{ij} - r_{ij})^2 \quad \text{for } r_{ij} < d_{ij}; \quad V(r_{ij}) = 0 \quad \text{otherwise.}$$

where k is the spring constant, $d_{ij} = d_i + d_j$ is the average diameter of the interacting particles. We have considered bidisperse systems, with a size ratio 1.4. We have prepared systems at different values of the pressure p , which is fixed to a high precision ($|dp|/p < 10^{-2}$) using a combination of energy minimization and compression/decompression steps. The final pressure spans 5 order of magnitudes, $p = 10^{-9} - 10^{-2}$.

C. Deformations

We measure local elastic constants by imposing small macroscopic compression or shear deformations to the above systems, respectively to measure local bulk and shear moduli. To explicitly check that we work in the linear response regime, we increase the strain through a sequence of small steps, minimizing the energy after each step through the conjugate gradient algorithm. We do observe the locally defined stress and strain to be linearly related, and thus estimate the local elastic constants

$$c_{ij}(\mathbf{r}, w) = \partial \sigma_i(\mathbf{r}, w) / \partial \varepsilon_j(\mathbf{r}, w),$$

through a linear best-fit of the stress versus strain relation. Here c_{ii} is to the local bulk modulus, while c_{ij} , with $i \neq j$, is the local shear modulus, under the assumption of rotational symmetry.

IV. RESULTS

A. Lennard Jones Systems

The local stress and the local strain are averages of particle and contact properties in the coarse graining region, as discussed in Sec. IIB, and therefore their s.d. is expected to scale as w^{-1} if w is larger than their correlation length. In the bidisperse LJ systems, we do actually observe the s.d. of the diagonal local stress, σ_{xx} , and the diagonal local strain, ε_{xx} , to scale as w^{-1} regardless of the fraction of large particles f_L , as illustrated in Fig. 1a,b. Conversely, the s.d. of the local off diagonal (shear) stress, σ_{xy} , and of the local off diagonal (shear) strain, ε_{xy} , scale as $w^{-\alpha}$, but α depends on f_L , as in panels d,e. Specifically, the exponent α is close to 1 only at high values of f_L . We also observe $\alpha \simeq 1$ for the weakly polydisperse LJ system, in agreement with earlier results [13].

A value $\alpha \neq 1$ implies the breakdown of the central limit theorem, and hence the presence of long-range correlations in the local shear stress and in the local shear strain [16, 17]. To explicitly check for the presence of these long-range correlations we investigate the correlation function of the local shear strain [16] ε_{xy} defined on the smallest of our investigated length scale, $w = 5$, for two values of f_L . Specifically, we have computed the

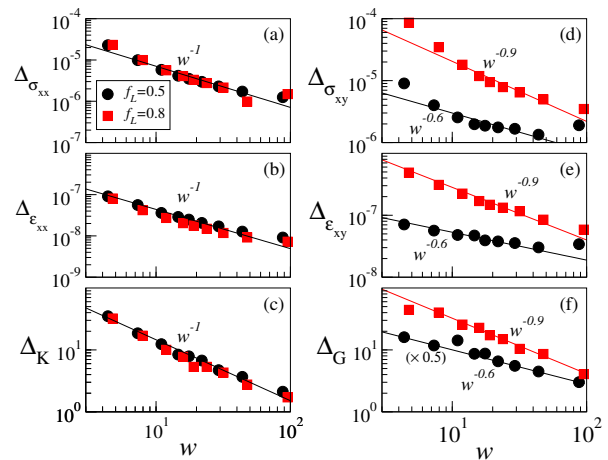


FIG. 1. Dependence of the s.d. of the local stress and of the local strain, and of the local elastic constants, on the coarse graining length scale. Panels a,b,c show results for the local diagonal strain, the local diagonal stress (pressure) and the local bulk modulus, while panels d,e,f show results for the local shear strain, the local shear stress and the local shear modulus. Data refer to the LJ bidisperse systems with fraction of large particles f_L , as indicated. For $f_L = 0.5$, data in panel f are scaled by 0.5 for clarity.

correlation map

$$C_{\varepsilon_{xy}}(x, y) = \frac{\langle \varepsilon_{xy}(0, 0) \varepsilon_{xy}(x, y) \rangle - \langle \varepsilon_{xy}^2(0, 0) \rangle}{\langle \varepsilon_{xy}(0, 0) \rangle^2 - \langle \varepsilon_{xy}^2(0, 0) \rangle}$$

and the correlations along the x -axis, the y -axis, and the diagonal, $C_{\varepsilon_{xy}}(r, 0)$, $C_{\varepsilon_{xy}}(0, r)$, and $C_{\varepsilon_{xy}}(r/\sqrt{2}, r/\sqrt{2})$ respectively, for $f_L = 0.8$ and 0.5 . For $f_L = 0.8$ we expect no long-range correlations as $\alpha \simeq 1$, and consistently observe the correlation functions to decay exponentially, as illustrated in Fig. 2a. On the contrary for $f_L = 0.5$ we expect long-range correlations. In this case the correlation functions exhibit persistent oscillations analogous to those recently experimentally observed in colloidal systems [16], as illustrated in Fig. 2b.

Local elastic constants are defined as the ratio of a local stress change and of a local strain change. Accordingly, as clarified by Eq. 4, their s.d. will asymptotically scale as that of the local stress or as that of the local strain, depending on which one asymptotically dominates. This is consistent with the results of Fig. 1c,f, where we show that the s.d. Δ_K of the local bulk modulus K scales as w^{-1} , while that of the local shear modulus scales as $w^{-\alpha}$, with α depending on f_L . To illustrate the connection between the exponents α_G , $\alpha_{\varepsilon_{xy}}$, $\alpha_{\sigma_{xy}}$, with which the s.d. of the local shear modulus, of the local shear strain and of the local shear stress scale with w , we show their dependence on f_L in Fig. 3(inset). Since $\alpha_{\varepsilon_{xy}} \leq \alpha_{\sigma_{xy}}$ at all f_L , the s.d. of the shear strain dominate that of the shear stress, and thus we expect from Eq. 4 $\alpha_G \simeq \alpha_{\varepsilon_{xy}}$, as observed.

The inset of Fig. 3 evidences that, in the bidisperse LJ systems, the exponents depend on the fraction of large

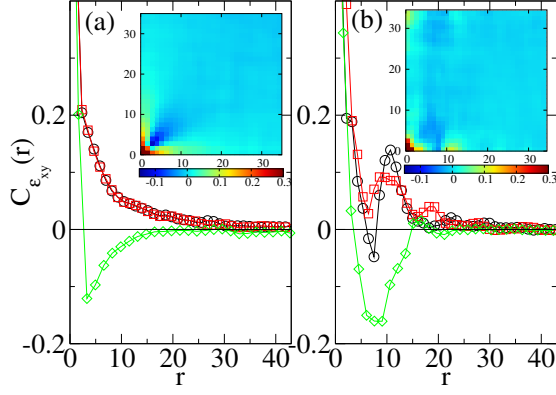


FIG. 2. Local shear strain correlations of bidisperse LJ systems with $f_L = 0.8$ (a), and $f_L = 0.5$ (b). The insets show the correlation map in the xy plane. The main panels illustrate the correlation along the x-axis (circles), the y-axis (squares), and the diagonal (diamonds). The size of the systems is $L \simeq 80$.

particles, f_L . This suggests that the exponents might generally depend on the ordering properties of the system. To check whereas this is the case we have investigated the degree of hexagonal ordering, which is the ordering expected to arise in 2d, evaluating the local orientational order parameter Q_m^{loc}

$$Q_m^{loc} = \frac{1}{N} \sum_{i=1}^{N_m} \frac{1}{n_i} \left| \sum_{j=1}^{n_i} e^{im\theta_{\mathbf{r}_{ij}}} \right|, \quad (5)$$

with $m = 6$. In the above expression, the first sum runs over all the particles, the second one over all n_i particles in contact with particle j , and $\theta_{\mathbf{r}_{ij}}$ is the angle formed by the vector \mathbf{r}_{ij} and a fixed arbitrary one, e.g. the x axis. We find the exponent α_G corresponding to different systems to collapse on a same master curve when plotted as a function of the order parameter, as in Fig. 3. Systems with a low degree of disorder, for which $Q_6^{loc} \simeq 1$, have no long range local shear strain or stress correlations, so that the s.d. of the local shear modulus scales as $w^{-\alpha}$ with $\alpha \simeq 1$. As the degree of disorder increases long range correlation emerges, and α decreases. If one assumes linear elasticity to hold to a good approximation above the length scale w_{LE} at which $\Delta_G(w_{LE})/\Delta_G(w_0) = x$, with x an arbitrary small threshold, w_0 a microscopic length scale, then one finds $w_{LE} \propto x^{-1/\alpha_G}$. Given the dependence of α_G on the disorder properties of the system, this implies that the length scale w_{LE} above which linear elasticity holds strongly increases as the degree of disorder increases.

B. Disk Particle Systems

Due to the approximate scale-invariance of the interaction potential, in LJ systems pressure plays the role

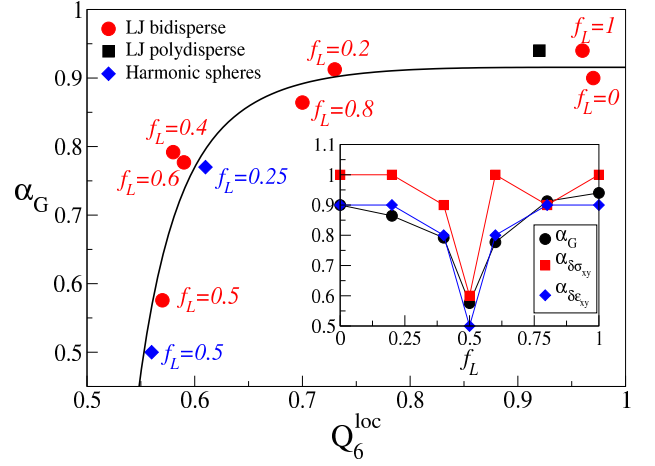


FIG. 3. The inset shows that the fraction f_L of large particle fixes the exponents governing the scaling of the s.d. of elastic properties with the coarse-graining length scale w , in the bidisperse LJ systems. Combining results obtained investigating different systems the main figure suggests that the exponent governing the scaling of the s.d. of the local shear modulus, α_G is fixed by the degree of disorder, as estimated by the local orientational order parameter Q_6^{loc} . The panel combines results of all three model systems we have considered. Bidisperse systems are investigated at different values of the fraction of large particles f_L .

of a stress scale. Accordingly, the pressure does not influence the ordering properties of the system, and hence the dependence of the s.d. of the local elastic constants on the coarse graining length scale. Conversely, in Harmonic systems close to the jamming transition ($p \rightarrow 0$) the macroscopic elastic constants scale critically with the pressure [24]. This makes of interest to investigate how the jamming transition influences the emergence of the continuum elastic limit.

The critical-like nature of the jamming transition suggests a scaling relation for Δ_G and Δ_K of the form,

$$\frac{\Delta_G(p, w)}{G(p)} = \mathcal{G} \left(\frac{w}{\xi_G(p)} \right); \quad \frac{\Delta_K(p, w)}{K(p)} = \mathcal{K} \left(\frac{w}{\xi_K(p)} \right), \quad (6)$$

where $G \propto p^{1/2}$ and $K \propto p^0$ close to the jamming transition [24].

We have verified these scaling relations investigating the dependence of the s.d. on w for Harmonic disks, and fraction large particles $f_L = 0.5$ and $f_L = 0.25$. Results for the bulk modulus are shown in Fig. 4a,c. The s.d. of the local bulk modulus asymptotically scales as w^{-1} , regardless of the value of f_L , and data corresponding to different pressures can be collapsed as suggested by the scaling relation of Eq. 6. The corresponding length scale ξ_K approaches a constant value in the $p \rightarrow 0$ limit, as illustrated in Fig. 5. Analogous results for the shear modulus are illustrated in Fig. 4b,d. In this case the asymptotic dependence of the s.d. on w depends on f_L , as in LJ systems, and the scaling of data corresponding to different pressures suggests $\xi_G \propto p^{-\nu_G}$ where $\nu_G \sim 0.25$, as

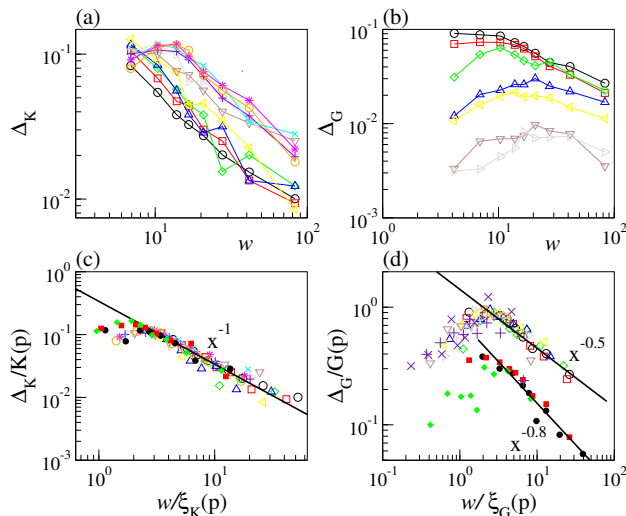


FIG. 4. Coarse graining length scale dependence of the s.d. of the local bulk modulus (a), for pressures in the range $p = 10^{-2} : 1.25 \times 10^{-7}$, from top to bottom, and of the shear modulus (b), for pressures in the range $p = 10^{-2} : 7 \times 10^{-5}$, from bottom to top. Panels c and d illustrate the collapse of these data according to Eq. 6, with $\xi_K \simeq \text{const.}$, and $\xi_G \propto p^{-0.25}$. All data refer to $f_L = 0.5$ system. In panel (d) we also illustrate data for $f_L = 0.25$ (full symbols, data scaled by 0.5), to clarify that the length scale ξ_G does not depend on f_L .

in Fig. 5. ξ_G can therefore be identified with the length scale ξ_c associated to the transverse vibrational modes of jammed systems, that exhibits the same pressure dependence in systems of Harmonic particles [22, 25]. Our findings for the scaling of Δ_K and Δ_G on p , and hence on the distance from the jamming threshold, are in line with recent results [14] obtained determining the local moduli using fluctuation formulas in the linear response regime [11, 12].

These results clarify that the pressure fixes the length scale above which the s.d. of the local elastic quantities scales as a power law of the coarse graining length scale. Conversely, the pressure does not influence the asymptotic behavior of the scaling functions, which behave as $\mathcal{G}(x) \propto x^{-\alpha_G}$, and as $\mathcal{K}(x) \propto x^{-1}$, respectively. The investigation of different values of f_L confirms the influence of the degree of order of the system on the value of the exponent α_G , as summarized in Fig. 3.

V. DISCUSSION

We have clarified that the process by which the continuum elastic limit emerges, as evaluated investigating the dependence of the standard deviation of the local elastic constants on the coarse graining length scale, is not universal. Rather, two properties of the system at hand influence the dependence of the s.d. of the local shear modulus with the coarse graining length scale. Asymp-

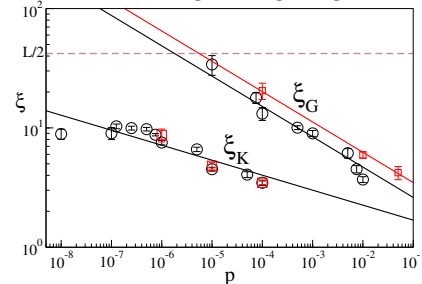


FIG. 5. Pressure dependence of the length scales $\xi_{K,G}$ for Harmonic system with different f_L . The solid lines represent power laws, $\nu_G = -0.25$. Squares (circles) show systems with $f_L = 0.25(0.5)$. We do not show data for ξ_G at smaller pressures, as they are affected by system size effects.

totically, the s.d. scales as a power law of the coarse graining length scale, with an exponent which is fixed by the degree of disorder. This is so as the local shear modulus is the ratio of a local shear stress and of a local shear strain, that could exhibit long range correlations in the presence of disorder. The power-law asymptotic regime occurs after a typical length scale, which is the typical length scale of the transverse vibrational modes. The role of this length scale is particularly relevant in systems of particles interacting via purely repulsive forces, as it diverges in the zero pressure limit corresponding to the jamming transition.

The main open question ahead is the individuation of the physical mechanisms responsible for the reported long range correlations in the elastic fields. There are contrasting results in the literature, as the local elastic moduli have been observed to be Gaussian in simulations of polymeric glasses [26] and of LJ glasses [13, 27], while the local shear strain has been observed to be long range in colloidal [16] and metallic [17] glasses, and theoretically predicted in granular systems [18]. Our results suggest that the investigation of the degree of disorder, or of other structural order parameter such as the excess entropy [28], could allow to rationalize this discrepancy.

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